

What Characteristics of Source Emissions can be used to Identify the Contribution of Different Source Types to Ambient PM Concentrations?

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Science Questions

This project aims to more fully characterize source emissions of PM_{2.5} and its gas-phase chemical precursors by addressing the following questions:

- -Which chemical compounds in PM_{2.5} may act as atmospheric emissions tracers for source apportionment modeling?
- Which source chemical signatures are useful to achieving a chemical mass balance solution for apportionment, attribution, and reconciliation?

Research Goals

Specifically, our investigations aim to:

- (1) sample several types of emissions sources with known environmental impacts;
- (2) measure PM_{2.5} mass and composition as a function of particles size in these emissions;
- (3) collect filter-based PM_{2.5} samples for analysis using state-of-the-art instrumentation;
- (4) develop new analytical techniques for the resolution of PM_{2.5} chemistry;
- (5) use the information generated above to develop chemical source signatures for use in source apportionment models; and
- (6) examine PM_{2.5} soot nanostructure and morphology in new ways.

Methods/Approach

Source selection is made on the basis of (i) the availability of a specific source's chemical emissions profile, (ii) the predominance of the emissions from that source, (iii) the potential environmental impacts of the source and (iv) the ability to gain on-site access to the facility, area, or equipment causing the emissions. Emissions are sampled using a dynamic dilution sampler (see Figure 1). Real-time particle size and PM_{2.5} mass distributions are measured, and filter-based samples are collected. PM_{2.5} filter samples are analyzed by various chemical methods. These methods identify and quantify the individual chemical constituents in PM_{2.5}. Application of a newly developed thermal extraction-GC-MS technique is also used to analyze impactor substrates to determine the individual chemical compound size-distributions. Lastly, a high resolution-transmission electron microscope is used to examine soot nanostructure and particle morphology.



Figure 1. Dynamic Dilution Sampler

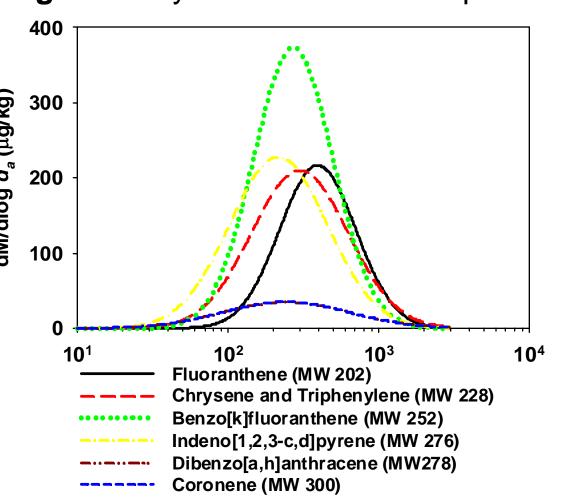


Figure 3. Size-dependent distribution of PAHs in wood smoke

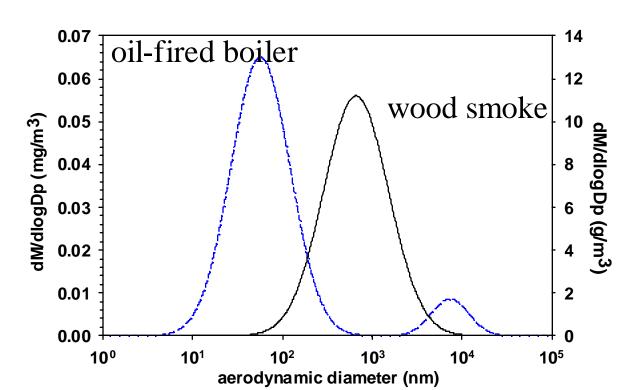


Figure 2. PM_{2.5} mass distributions from residential wood combustion and oil-fired boiler appliances

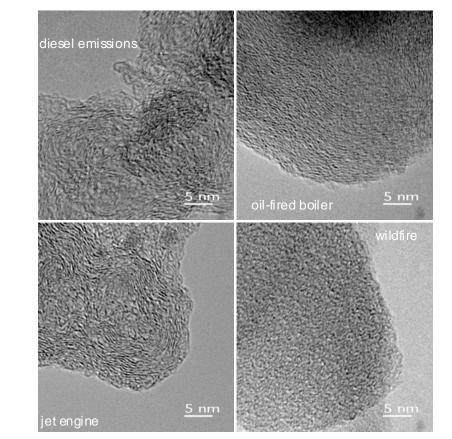


Figure 4. HR-TEM images of various source emissions

Results/Conclusions

The source characterization program has sampled emissions from wildfires, residential wood combustion, agricultural burns, diesel vehicle exhaust, jet engine exhaust, and large-scale industrial stacks (e.g. oil-fired boiler, biomass boilers, pulp and paper mill). The mass and chemistry of PM_{2.5} is source dependent. For example, emissions from burning biomass tend to comprise large concentrations of organic carbon, whereas diesel vehicle and oil-fired boiler samples are composed of mostly elemental carbon. Certain individual species are source-specific.

Figure 2 shows that the size distribution of PM_{2.5} mass can vary by source. The residential oil-fired boiler emissions emit in the ultra-fine mode (<100nm), whereas wood smoke is generally in an accumulation mode. The small peak in the larger particle diameter region of Figure 2 is likely agglomerates of primary oil-fired boiler particles.

Figure 3 shows that some individual organics in fine PM from wood smoke are concentrated in particles of smaller geometric mean diameters compared to the PM mass (compare Figs. 2 and 3). Understanding size-specific speciation is critical to advancing future generation dispersion models and linking source emissions to health end points.

Figure 4 illustrates the application of high-resolution transmission electron microscopy to several field-collected PM filter samples. These nanostructures appear to be source specific and may prove to be a useful tool in furthering source apportionment and attribution research.

Future Directions

- Create a source database containing size distributions of individual chemical species for future generation dispersion models.
- Develop chemical profiles of high-molecular weight species in fine PM for use in source apportionment, for a better understanding of the organic carbon fraction, and to achieve a PM_{2.5} mass balance.
- Continue to study the use of high resolution imaging for definition of soot nanostructure in fine PM and for source apportionment.

Impact and Outcomes

- New chemical profiles have been added to SPECIATE database for use by EPA and state regulatory offices. This results in more accurate allocation of contributions from different source types, and thus the development of reduction strategies that target the sources contributing the most to excessive ambient PM concentrations.
- Our source data are being used in sourcereceptor models developed to study air quality at the PM Supersites.
- Our source data are being used to explain regional air pollution episodes that have resulted in non-compliance with NAAQS.
- Our developed thermal extraction method has been adopted by several PM research laboratories.
- Improved data on size-dependent speciation allows more accurate dispersion modeling and subsequently more accurate estimates of exposure to potentially harmful PM components.

